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TITLE: Process for preparing a carbonaceous five-layer fuel cell electrode substrate with elongated holes for feeding reactant gases

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CLAIMS:

What is claim is:
1. A process for forming an electrode substrate for fuel cells comprising the steps of:
(1) supplying in order, to a mold having a predetermined configuration:
(a) a material for a first porous carbonaceous layer,
(b) a material for forming first elongated holes,
(c) a material for a first dense carbonaceous layer having a bulk density greater than a bulk density of the first porous layer,
(d) a material for a separator,
(e) a material for a second dense carbonaceous layer,
(f) a material for forming second elongated holes perpendicular to the first elongated holes, and
(g) a material for a second porous carbonaceous layer having a bulk density less than a bulk density of the second dense layer;
(2) press molding the materials in the mold;
(3) post-curing the press-molded material; and
(4) calcinating the post-cured material in an inert atmosphere.
2. A process for forming an electrode substrate for fuel cells comprising the steps of:
(1) supplying, in order, a material for a first dense carbonaceous layer, a material for forming first elongated holes, and a material for a first porous carbonaceous layer having a bulk density less than a bulk density of the first dense layer into a mold having a predetermined configuration;
(2) pre-pressing the materials in the mold so as to form a first pre-pressed product;
(3) removing the first pre-pressed product from the mold;
(4) supplying, in order, a material for a second dense carbonaceous layer, a material for forming second elongated holes, and material for a second porous carbonaceous layer having a bulk density less than a bulk density of the second dense layer into a mold with a predetermined configuration;
(5) pre-pressing the materials in the mold so as to form a second pre-pressed product;
(6) removing the second pre-pressed product from the mold;
(7) placing the first pre-pressed into a mold with the first porous layer

facing downwardly;
(8) supplying a material for a separator to the mold;
(9) placing the second pre-pressed product with the second dense layer facing the material for the separator and with the second elongated holes disposed perpendicular to the first elongated holes;
(10) press molding the materials in the mold;
(11) post-curing the press-molded material; and
(12) calcinating the post-cured material in an inert atmosphere.
3. The process of claim 1, wherein the steps of supplying a material for a first porous layer and a second porous layer comprises supplying a mixture comprising about 10 to about 50% by weight of a filler, about 20 to about 40% by weight of a binder, and about 20 to about 50% by weight of a pore regulator.

4. The process of claim 3, wherein the steps of supplying a material for the first and second porous layers further comprises selecting the filler from the group consisting of short carbon fibers and carbon particles.
5. The process of claim 4, wherein the step of selecting the filler comprises selecting short carbon fibers having a diameter in the range of from about 5 to about 30 microns, a length in the range of from about 0.02 to about 2.0 mm, and a linear carbonizing shrinkage in the range of not more than about 3.0% when calcinated at 2000.degree. C.
6. The process of claim 3, wherein the step of supplying a material for the first and second porous layers further comprises selecting a binder from the group consisting of a phenol resin, an epoxy resin, a petroleum pitch, a coal pitch and mixtures thereof, and has a carbonizing yield in the range of from about 30 to about 75% by weight.
7. The process of claim 3, wherein the step of supplying a material for the first and second porous layers further comprises selecting a pore regular comprising organic granules, about 70% or more of which have a particle diameter in the range of from about 30 to about 300 microns.
8. The process of claim 7, wherein the step of selecting a pore regulator further comprises selecting the organic granule from the group consisting of polyvinyl alcohols, polyvinyl chlorides, polyethylenes, polypropylenes, polystyrenes, and mixtures thereof.
9. The process of claim 1, wherein the step of supplying a material for forming first and second elongated holes comprises supplying a polymer.
10. The process of claim 9, wherein the step of supplying a polymer comprises supplying a polymer that does not evaporate nor melt-flow at 100.degree. C.
11. The process of claim 10, wherein the step of supplying a polymer further comprises selecting a polymer from the group consisting of polyethylenes, polypropylenes, polystyrenes, polyvinyl alcohols and polyvinyl cholorides, and having a carbonizing yield of about 30% by weight or less.
12. The process of claim 9, wherein the step of supplying a polymer comprises supplying at least one of a textile fabric and a grating-like shaped article of polymer.
13. The process of claim 12, wherein the step of supplying a polymer comprises supplying a textile fabric which comprises at least one of single strands and bundles of a number of strands which have been textured; said single strands and said bundles each having a diameter in the range of about 0.5 to about 3.3 mm.
14. The process of claim 13, wherein the step of supplying a textile fabric further comprises placing said strands and said bundles such that adjacent strands and adjacent bundles parallel to the gas flow direction are spaced apart a distance in the range of from about 1.5 to about 5.0 mm and such that the distance between adjacent strands and adjacent bundles perpendicular to the gas flow direction is in the range of from about 5 to about 50 mm.
15. The process of claim 12, wherein the step of supplying a material for forming the first and second elongated holes comprises supplying a grating-like shaped article of polymer prepared by one of extrusion molding of a melt of the polymer into a mold, press molding of pellets in a mold and press molding of powder in a mold and the grating-like shaped article has a diameter in the range of from about 0.5 to about 3.3 mm.
16. The process of claim 15, wherein the step of supplying the grating-like shaped article comprises positioning the grating-like shaped article such that adjacent gratings parallel to the gas flow direction are spaced apart a distance in the range of from about 1.5 to about 5.0 mm and the distance between adjacent gratings perpendicular to the gas flow direction is in the range of from about 5 to about 50 mm.
17. The process of claim 1, wherein the step of supplying a material for a

first dense layer and a second dense layer comprises supplying a mixture comprising about 30 to about 70% by weight of a filler, about 20 to about 40% by weight of a binder, and about 10 to about 30% by weight of a pore regulator.

18. The process of claim 1, wherein the step of supplying a material for a separator comprises supplying one of a carbon plate and a graphite sheet.
19. The process of claim 1, wherein the step of supplying a material for a separator comprises supplying a powdery mixture comprising a filler selected from the group consisting of short carbon fibers, fine powders of carbonaceous precursor and carbon particles, and a binder selected from the group consisting of a phenol resin, an epoxy resin and a furan resin.
20. The process of claim 1, wherein the step of press molding comprises press molding at a temperature in the range of from about 70 to about 170.degree. C. and a pressure in the range of from about 5 to about 100 kg/cm.sup.2 for a time period in the range of from about 1 to about 60 minutes.
21. The process of claim 1, wherein the step of post-curing comprises post-curing at the molding temperature for at least about 2 hours.
22. The process of claim 1, wherein the step of calcination comprises calcinating under an inert atmosphere at a temperature in the range of from about 800.degree. to about 3000.degree. C. for about 1 hour.
23. The process of claim 2, wherein the steps of pre-press molding comprise pre-press molding at a temperature in the range of from about 60.degree. to about 100.degree. C. and a pressure in the range of from about 20 to about 50 kg/cm.sup.2 for a time period in the range of from about 10 to 30 minutes.
24. The process of claim 2, wherein the step of press molding comprises press molding at a temperature in the range from about 120.degree. to about 160.degree. C. and a pressure in the range of from about 20 to about 50 kg/cm.sup.2 for a time period in the range of from about 10 to about 30 minutes.